# The Charge Ordered State from Weak to Strong Coupling

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## Abstract

We apply the Dynamical Mean Field Theory to the problem of charge ordering. In the normal state as well as in the Charge Ordered (CO) state the existence of polarons, i.e. electrons strongly coupled to local lattice deformation, is associated to the qualitative properties of the Lattice Polarization Distribution Function (LPDF). At intermediate and strong coupling a CO state characterized by a certain amount of thermally activated defects arise from the spatial ordering of preexisting randomly distributed polarons. Properties of this particular CO state gives a qualitative understanding of the low frequency behavior of optical conductivity of $Ni$ perovskites.

**KEY WORDS:** Charge Ordering,Polarons

## 1. INTRODUCTION

Recently there has been a renewed interest for the charge ordering transition which has been found associated to lattice displacements in Cuprates Nickelates and Manganites.

Charge stripes order has been detected in Neodimium doped Cuprates ($La_{1.475}Nd_{0.4}Sr_{0.125}CuO_4$) [1] and conjectured in $LASCO$ [2]. X ray studies of $BISCO$ also shown a modulated structure of $CuO$ planes [3]. Commensurate charge order appears in doped Nickelates [2] also related with peculiar magnetic properties [4], and finally large lattice distortions have been found in Manganites which can be associated to either commensurate or incommensurate charge ordering [5]. The amplitude of such a distortion increases from Cuprates to Manganites, a fact that may support the hypothesis of an increasing charge-lattice interaction. Another important observation supporting the presence of polaronic carrier, and therefore an intermediate-strong local charge-lattice interaction, is the presence of a polaronic peak in the MIR band of doped $Ni$ compound [6] [7].

The aim of this paper is to show how non-perturbative results obtained in the framework of the Dynamical Mean Field Theory (DMFT) can be helpful to understand the low energy behavior of the optical conductivity in $Ni$ perovskites. We shall first summarize the results of our theory of the charge ordered (CO) state, then we will show a calculation of the optical conductivity which is in qualitative agreement with experimental observation in

charge ordered $Ni$ perovskites. In the weak coupling case the CO transition is the well known Charge Density Wave instability of the Fermi liquid. While in strong coupling there have been several attempts to understand the CO ordered state based on mean field approach on strong coupling effective Hamiltonians [8] and studies of the ground state [9].

To discuss the CO state in a non perturbative fashion we consider the simplest model of local electron lattice interaction i.e. the Holstein molecular crystal model [10]

$$H = \sum_i \frac{P_i^2}{2M} + \frac{1}{2}kX_i^2 - \frac{t}{2\sqrt{z}}\sum_{ij} c_i^\dagger c_j + g\sum_i (c_i^\dagger c_i - n)X_i \tag{1}$$

where $c_i^\dagger(c_i)$ creates (destroys) an electron at site $i$, and $X_i$, $P_i$ are the local oscillators displacements and momentum. Electrons and phonons are coupled via the density fluctuations ($n$ being the average electron density). The electrons move on a bipartite lattice of connectivity $z$ and have a band of half-bandwidth $t$.

The main approximation we consider is the adiabatic approximation which can be obtained in the limit $M \to \infty$. In this limit we neglect the first term in eq. (1) therefore $X_i$ are constant of motion and can be replaced by $c$-numbers. This approximation turns out to be valid if the following two conditions hold

i) as far as thermodynamic properties are concerned temperatures must be greater than the typical phonon energy scale $\omega_0 = \sqrt{k/M}$ [11]

ii) as far as spectral properties are concerned energies must be greater than the typical phonon energy scale $\omega_0$

On the other hand adiabatic limit allows to solve the model with a little amount of numerics giving the spectral properties of electrons in *real frequencies* and statistical properties of the lattice. We consider also *spinless electron* to account for a polaronic rather than a bipolaronic ground state at large couplings. This restriction even if at a very rough level, mimics the action of an on site Coulomb repulsion.

We will apply the machinery of the Dynamical Mean Field Theory which is the exact solution of local-type interaction on an infinite coordination lattice $z \to \infty$ (infinite dimensions) [14]. To have a non trivial limit a scaling of the hopping, as in eq. (1), $t$ with the number of neighbors is required. The DMFT approach maps the problem of locally interacting fermions on a lattice into a single site equivalent problem [14]. A detailed study of the Holstein model based on the Montecarlo solution of the single site problem has been first carried out in ref. [15]. This analysis has been extended to the spectral properties of the normal state in ref. [11] by using the adiabatic limit to obtain an analytical solution of the single site model. We extend this analytical approach to the study of the charge ordered (CO) state. We consider here alternate charge ordering in two interpenetrating sublattices $A$ and $B$. The quantity to be determined self-consistently is the Lattice Polarization Distribution Function (LPDF) $P(X)$. Different regimes are related to qualitative changes in the shape of $P(X)$. Our main results can be summarized by the self-consistent equations which determines the LPDF and the local electron Green function in each sublattice

$$P_{A,B}(X) \propto e^{-kX^2/2}\pi_n\left(i\omega_n - \frac{t^2}{4}G_{B,A}(i\omega_n) - gX_{A,B}\right) \tag{2}$$

$$G_{A,B}(i\omega_n) = \int dX P_{A,B}(X)\frac{1}{i\omega_n - \frac{t^2}{4}G_{B,A}(i\omega_n) - gX} \tag{3}$$

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Equations (2,3) are obtained in the simple but non trivial case of Bethe lattice of bandwidth $2t$. From eq. (3) we see that the Green function is that of a particle propagating in a randomly distorted sublattice and sublattice $A$ is coupled to $B$ and vice versa. The real frequency representation of the (retarded) Green function is simply obtained substituting $i\omega_n \rightarrow \omega + i0^+$ in eq. (3), therefore the adiabatic limit allows to obtain the spectral properties in real frequencies.

## 2. RESULTS

We have solved the self-consistent scheme introduced in the previous section by numerical iteration procedure. We consider the spinless electron half filled case i.e. one electron each two sites ($n = 0.5$). We start with an ansatz for the sublattice Green function then we get the function $P$ at discrete points trough eq. (2) and trough a numerical integration (eq. (3) we obtain the new $G$.

In the adiabatic limit only one relevant coupling parameter measures the electron-lattice interaction $\lambda = g^2/2kt$. It can be expressed as the ratio of self-trapping energy (polaron energy $\epsilon_p = g^2/2k$) to the electron kinetic energy energy ($t$). A crossover from strong to weak coupling behavior is expected around $\lambda \simeq 1$ [12,13]. These expectations are confirmed by the the phase diagram at half filling shown in fig. . The continuous curve represents the CO critical temperature as a function of the coupling strength. The dashed line marks the normal to polaron crossover in the normal phase [11] and the crossover from weak coupling CO (A) to strong coupling CO (B). In both normal and ordered state a crossover line separates the monomodal and the bimodal behavior of the LPDF, in the ordered state bimodality appears in the sublattice LPDF. Results are summarized in fig. . The typical weak coupling behavior of LPDF across the transition temperature is shown in fig. a). Upon decreasing the temperature an uniform polarization of a given sublattice arises. The other sublattice, whose LPDF is not shown in the figure, develops an opposite polarization so that the net total polarization is zero as should be for the hamiltonian eq. (1) which couples deformation and density fluctuations. The variance decreases with decreasing temperature.

Moving to polaronic non ordered state the LPDF is clearly bimodal and symmetric at half filling as it is seen from fig. c) (dashed lines). Upon decreasing the temperature below $T_c$ the sublattice LPDF unbalances in favor of a net sublattice polarization but still remain bimodal. The weight of the secondary peak decreases by a further decreasing of temperature. We explain this secondary peak as due to temperature activated *defects* in the CO ordered state [9].

The bimodal behavior of LPDF which is clear at very large coupling becomes less pronounced at intermediate coupling (see fig. b)). In this region in both non ordered and ordered state near $T_c$ we observe that a non negligible amount of sites are nearly undistorted. In this intermediate region of the coupling $0.69 > \lambda > 0.59$ we observe a strong non Gaussian behavior of LPDF. Even if a secondary peak is present in the normal phase it is not well pronounced in this region. As the temperature is lowered below $T_c$ it may happen that this secondary peak is washed out in the ordered phase but a pronounced shoulder remains in the LPDF. It eventually develops again a secondary peak upon a further decrease in temperature.

The Optical Conductivity is obtained once the local Green functions of the two sublattices are known by a generalization of the Kubo formula.

Details of the calculation will be presented elsewhere. We show in fig. the results obtained at half filling for three different values of the coupling constant characteristic of small, intermediate and large couplings. We see that at small coupling (fig. a)) the optical conductivity of the normal state shows a peak at $\omega \simeq 0$ reminiscent of the classical Drude behavior. This peak is shifted in the CO state at $\omega = 2\Delta$ where $\Delta$ is the CDW gap. The position of the peak depends on temperature and is shifted toward higher energies as the temperature decreases following the enhancing of the order parameter. As the coupling is increased (fig. b)) a peak at $\omega > 0$ *is still present* also at $T > T_c$ indicating the presence of polarons in the disordered phase. We notice however a shift of the spectral weight from low to high energies as the temperature is decreased. This effect is less evident at stronger couplings ( c)) because in this case almost all sites are polarized (see. fig. c)) and consequently we have no appreciable spectral weight at low energy. In any case whenever polarons are present in the non ordered state a shift toward larger energies of the spectral weight and a change in the temperature dependence of the amplitude of the peak is observed in the ordered state.

A strong coupling approximation, equivalent to Reik's approximation [16] at high temperatures [17], gives the interpretation of this spectral weight shift in terms of the number of defects in the CO state. In this approximation the ratio of the optical conductivity in the CO state to the same quantity in the normal state is

$$\frac{\sigma_{CO}(\omega)}{\sigma_{norm}(\omega)} = 1 - (1/2 - n_d)^2 F(T, \omega). \tag{4}$$

In eq. (4) $n_d$ is number of defects in the charge ordered state which depends on $T - T_c$ whereas $F(T, \omega)$ is a function of $T$ only

$$F(T, \omega) = \sqrt{\beta/2\pi\epsilon_p} \int d\nu \; e^{-\nu^2/2\epsilon_p} \times tanh(\frac{\beta(\nu - \omega/2)}{2}) tanh(\frac{\beta(\nu + \omega/2)}{2}) \tag{5}$$

. From this formula it is easy to obtain a spectral weight shift proportional to $(1 - 2n_d)^2$ from low to high energy.

It is worth to note that the shift of the spectral weight from low to high energy is a phenomenon actually observed in the $Ni$ oxides together with an anomalous ratio $2\Delta(0)/T_c = 13$ [7]. Both observations are consistent with an intermediate coupling charge ordering of polarons.

## 3. CONCLUSIONS

The crossover from weak to strong coupling CO have been studied in details by introducing a LPDF. The qualitative change from monomodal to bimodal behaviour of this function is interpreted as existence of defects in the ordered phase. In terms of the defects activation we obtain a qualitative understanding of the shift from low to high frequency in the spectral weight observed below the CO transition in $Ni$ perovskites.

## ACKNOWLEDGEMENTS

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## Figure caption

Fig. 1 The phase diagram at half-filling. Dots are the data obtained from numerical solution of eqs. (2,3), dashed line marks the separation from unimodal LPDF to bimodal LPDF in both normal and CO state.

Fig. 2 LPDF of the sublattice $A$ above (dashed) and below (solid $T_c$) for a) $\lambda = 0.4$ b) $\lambda = 1$ c) $\lambda = 2$. Temperature ranges are a) from $T_{in} = 5 \times 10^{-3}$ to $T_{fin} = 3 \times 10^{-2}$ with temperature step $\Delta T = 5 \times 10^{-3}$, b) $T_{in} = 2 \times 10^{-2}$, $T_{fin} = 6 \times 10^{-2}$, $\Delta T = 5 \times 10^{-3}$, c) $T_{in} = 2 \times 10^{-2}$, $T_{fin} = 5 \times 10^{-2}$, $\Delta T = 2.5 \times 10^{-3}$. Temperature is in units of the half-bandwidth $t$.

Fig. 3 Optical conductivity above (dashed) and below (solid $T_c$) for a) $\lambda = 0.4$ b) $\lambda = 1$ c) $\lambda = 2$. Temperature ranges are the same as in fig.